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Supplementary Materials for

Like dissolves like: A first-principles theory for predicting liquid miscibility and mixture dielectric constant

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S1. DERIVATION OF THE THEORY

A. The set-up

Here, we describe the detailed derivation of the theory. We consider a fluid mixture at uniform density in an applied field $\mathbf{E}_0(\mathbf{r})$. For each species S in the fluid, there are N_s molecules. The fluid molecules are modeled as nonpolarizable, each having a permanent dipole moment $\bar{\mu}_s$ and volume v_s. A microscopic state of the fluid can be specified by the set of positions and the dipole vectors of all molecules $\{(\mathbf{r}_{S,i}, \boldsymbol{\mu}_{S,i})\}$, where $\mathbf{r}_{S,i}$ and $\boldsymbol{\mu}_{S,i}$ ($|\boldsymbol{\mu}_{S,i}| = \bar{\mu}_S$) are the center-of-mass position and the dipole moment of the ith molecule of type S.

To describe the electrostatic energy of the fluid mixture, we first introduce a microscopic polarization $\hat{\mathbf{P}}(\mathbf{r})$, given by

$$\hat{\mathbf{P}}(\mathbf{r}) = \sum_S \sum_{i=1}^{N_S} \boldsymbol{\mu}_{S,i} h_S(\mathbf{r} - \mathbf{r}_{S,i}) \tag{S1}$$

where $h_S(\mathbf{r} - \mathbf{r}_{S,i})$ is a function describing the local spread of the molecular polarization around the center of mass of the molecule. In terms of $\hat{\mathbf{P}}(\mathbf{r})$, the electrostatic energy of the fluid is

$$U = \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \, \hat{\mathbf{P}}(\mathbf{r}) \mathbf{T}(\mathbf{r} - \mathbf{r}') \hat{\mathbf{P}}(\mathbf{r}') - \int d\mathbf{r} \, \hat{\mathbf{P}}(\mathbf{r}) \cdot \mathbf{E}_0(\mathbf{r}) \tag{S2}$$

where $\mathbf{T}(\mathbf{r}) = -\nabla\nabla \left(1/4\pi\varepsilon_0 |\mathbf{r}|\right)$ is the dipole-dipole interaction tensor. Mathematically, the improper integrals involving $\mathbf{T}(\mathbf{r})$ is known to be nonunique. However, the problem is well understood[38,39] and a consistent interpretation of $\mathbf{T}(\mathbf{r})$ is given by the following expression[40]:

$$\begin{aligned} \mathbf{T}(\mathbf{r}) &= -H(|\mathbf{r}| - \eta) \nabla\nabla \frac{1}{4\pi\varepsilon_0 |\mathbf{r}|} + \frac{1}{3\varepsilon_0} \mathbb{1} \delta(\mathbf{r}) \\ &= H(|\mathbf{r}| - \eta) \left(\frac{1}{4\pi\varepsilon_0 |\mathbf{r}|^3} \left[\mathbb{1} - \frac{3\mathbf{r}\mathbf{r}}{|\mathbf{r}|^2} \right] \right) + \frac{1}{3\varepsilon_0} \mathbb{1} \delta(\mathbf{r}) \end{aligned} \tag{S3}$$

where $\mathbb{1}$ is the unit dyadic, $\delta(\mathbf{r})$ is the Dirac delta function, and $H(x)$ is the Heaviside step function. η is a regularization parameter which will be taken to 0 eventually. Since both terms in $\mathbf{T}(\mathbf{r})$ are well defined, it is now straightforward to compute the Fourier transform of $\mathbf{T}(\mathbf{r})$, and the result is[35,40]

$$\widetilde{\mathbf{T}}(\mathbf{k}) = \frac{\mathbf{k}\mathbf{k}}{\varepsilon_0 k^2} \tag{S4}$$

where a tilde above a quantity denotes the Fourier transform $\tilde{f}(\mathbf{k}) = \int d\mathbf{r} \, f(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}}$ and $f(\mathbf{r}) = \frac{1}{(2\pi)^3} \int d\mathbf{k} \tilde{f}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}}$. Noting that $\mathbf{k}\mathbf{k}/k^2$ is a projection operator, we have the following relation

$$\left(\varepsilon_0 \widetilde{\mathbf{T}}(\mathbf{k}) \right)^n = \varepsilon_0 \widetilde{\mathbf{T}}(\mathbf{k}) \tag{S5}$$

which turns out to be useful in the algebraic manipulations in this work.

We consider a grand canonical ensemble of the fluid mixture under chemical potential μ_S for each species S at temperature T and volume V. The grand partition function of the system is given by

$$\Xi = \sum_{N_A=0}^{\infty} \cdots \sum_{N_B=0}^{\infty} \frac{e^{\beta\mu_A N_A}}{N_A!} \cdots \frac{e^{\beta\mu_B N_B}}{N_B!} Z(\{N_S\}) \tag{S6}$$

where A, B are representative labels of the solvent species and the ... means similar summations and factors for other species need to be included if there are more than two species in the mixture. $Z(\{N_S\})$ is the canonical partition function given by

$$Z(\{N_S\}) = \prod_S \prod_{i=1}^{N_S} \int \frac{d\mathbf{r}_{S,i}}{\Lambda_S^3} \int \frac{d\Omega_{S,i}}{4\pi} e^{-\beta U} \tag{S7}$$

where Λ_S is the thermal deBroglie wavelength of species S and the integral of the dipole moment is carried out over the solid angle $\Omega_{s,i}$ of the permanent dipole moments.

B. The transformation to field-based partition function

Next, we use the Faddeev-Popov method[31–33] to decouple the pairwise dipole-dipole interactions. This is done by introducing the δ-functional, which is the generalization of the multivariate δ-function:

$$\mathbf{1} = \int \mathcal{D}\mathbf{P}\, \delta[\mathbf{P}(\mathbf{r}) - \hat{\mathbf{P}}(\mathbf{r})] = \int \mathcal{D}\mathbf{P} \int \mathcal{D}\boldsymbol{\mathcal{G}}\, e^{i\int d\mathbf{r}\, \boldsymbol{\mathcal{G}}(\mathbf{r})\cdot[\mathbf{P}(\mathbf{r}) - \hat{\mathbf{P}}(\mathbf{r})]} \tag{S8}$$

We insert the δ-functional into the canonical partition function in Eq. (S6). This allows us to rewrite the Boltzmann factor as

$$e^{-\beta U} = \int \mathcal{D}\mathbf{P} \int \mathcal{D}\boldsymbol{\mathcal{G}}\, e^{-\frac{1}{2}\int d\mathbf{r}\int d\mathbf{r}' \mathbf{P}(\mathbf{r})\boldsymbol{\mathcal{T}}(\mathbf{r}-\mathbf{r}')\mathbf{P}(\mathbf{r}') - i\int d\mathbf{r}\, \mathbf{P}(\mathbf{r})\cdot\boldsymbol{\mathcal{G}}(\mathbf{r}) + \int d\mathbf{r}\, \mathbf{P}(\mathbf{r})\cdot\boldsymbol{\mathcal{E}}_0(\mathbf{r}) + i\int d\mathbf{r}\, \hat{\mathbf{P}}(\mathbf{r})\cdot\boldsymbol{\mathcal{G}}(\mathbf{r})} \tag{S9}$$

In the above expression, we have defined $\boldsymbol{\mathcal{T}} = \beta\mathbf{T}$ and $\boldsymbol{\mathcal{E}}_0 = \beta\mathbf{E}_0$ to simplify the expression. We note that in the exponent on the right-hand-side of Eq. (S9), only the last term depends explicitly on the instantaneous molecular configuration. By rearranging the order of the integrals in the canonical partition function, we can rewrite $Z(\{N_S\})$ in the following form:

$$Z(\{N_S\}) = \int \mathcal{D}\mathbf{P} \int \mathcal{D}\boldsymbol{\mathcal{G}}\, e^{-\frac{1}{2}\int d\mathbf{r}\int d\mathbf{r}' \mathbf{P}(\mathbf{r})\boldsymbol{\mathcal{T}}(\mathbf{r}-\mathbf{r}')\mathbf{P}(\mathbf{r}') - i\int d\mathbf{r}\, \mathbf{P}(\mathbf{r})\cdot\boldsymbol{\mathcal{G}}(\mathbf{r}) + \int d\mathbf{r}\, \mathbf{P}(\mathbf{r})\cdot\boldsymbol{\mathcal{E}}_0(\mathbf{r})} \prod_S q_S[\boldsymbol{\mathcal{G}}]^{N_S} \tag{S10}$$

where $q_S[\boldsymbol{\mathcal{G}}]$ is the single-dipole partition function under the fluctuating field $\boldsymbol{\mathcal{G}}$:

$$q_S[\boldsymbol{\mathcal{G}}] = \frac{1}{4\pi\Lambda_S^3} \int d\mathbf{r} \int d\Omega_S\, e^{i\int d\mathbf{r}' h_S(\mathbf{r}'-\mathbf{r})\boldsymbol{\mu}_S\cdot\boldsymbol{\mathcal{G}}(\mathbf{r}')} \tag{S11}$$

Substituting the field-based expression for $Z\{N_S\})$ into the grand partition function Ξ, we write the field-based expression for Ξ as:

$$\Xi = \int \mathcal{D}\mathbf{P} \int \mathcal{D}\boldsymbol{\mathcal{G}}\, e^{-L[\mathbf{P},\boldsymbol{\mathcal{G}}]} \tag{S12}$$

where the effective field-theoretic action L is

$$\begin{aligned} L[\mathbf{P},\boldsymbol{\mathcal{G}}] = {} & \frac{1}{2}\int d\mathbf{r} \int d\mathbf{r}' \mathbf{P}(\mathbf{r})\boldsymbol{\mathcal{T}}(\mathbf{r}-\mathbf{r}')\mathbf{P}(\mathbf{r}') + i\int d\mathbf{r}\, \mathbf{P}(\mathbf{r})\cdot\boldsymbol{\mathcal{G}}(\mathbf{r}) - \int d\mathbf{r}\mathbf{P}(\mathbf{r})\cdot\boldsymbol{\mathcal{E}}_0(\mathbf{r}) \\ & - \sum_S \left(\lambda_S \int d\mathbf{r} \int d\Omega_S\, e^{i\int d\mathbf{r}'\, h_S(\mathbf{r}'-\mathbf{r})\boldsymbol{\mu}_S\cdot\boldsymbol{\mathcal{G}}(\mathbf{r}')}\right) \end{aligned} \tag{S13}$$

where $\lambda_s = e^{\beta\mu_S}/(4\pi\Lambda_S^3)$ is the scaled fugacity of species S.

C. The variational approach

We approximate the grand potential of the system using the variational approach introduced in Eqs. (17)–(20) of the main manuscript. To evaluate the grand potential using Eq. (17), we have to evaluate Ξ_0 and $\langle L - L_0 \rangle_0$. We shall first evaluate Ξ_0. Using standard techniques for Gaussian integrals, we arrive at the following expression for Ξ_0:

$$\Xi_0 = \left(\frac{\det \boldsymbol{\mathcal{A}}}{\det(\boldsymbol{\mathcal{T}} + \boldsymbol{\mathcal{A}})} \right)^{\frac{1}{2}} \exp\left\{ \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \left[\boldsymbol{\mathcal{E}}_0(\mathbf{r}) - \boldsymbol{\mathcal{F}}(\mathbf{r}) \right] \left[\boldsymbol{\mathcal{T}}(\mathbf{r} - \mathbf{r}') + \boldsymbol{\mathcal{A}}(\mathbf{r} - \mathbf{r}') \right]^{-1} \left[\boldsymbol{\mathcal{E}}_0(\mathbf{r}') - \boldsymbol{\mathcal{F}}(\mathbf{r}') \right] \right\} \tag{S14}$$

where the determinant of an operator is defined as

$$\begin{aligned} (\det \boldsymbol{\mathcal{M}})^{-\frac{1}{2}} &= \int \mathcal{D}\boldsymbol{\xi} \, \exp\left[-\frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \boldsymbol{\xi}(\mathbf{r}) \boldsymbol{\mathcal{M}}(\mathbf{r} - \mathbf{r}') \boldsymbol{\xi}(\mathbf{r}') \right] \\ &= \int \mathcal{D}\boldsymbol{\xi} \, \exp\left[-\frac{1}{2} \int \frac{d\mathbf{k}}{(2\pi)^3} \tilde{\boldsymbol{\xi}}(\mathbf{k}) \widetilde{\boldsymbol{\mathcal{M}}}(\mathbf{k}) \tilde{\boldsymbol{\xi}}(-\mathbf{k}) \right] \end{aligned} \tag{S15}$$

To evaluate the term involving the determinants, we use a strategy similar to that described in Appendix B of Ref. 41. The evaluation of the term is described in detail in Eqs. (B4) – (B8) in our earlier manuscript in Ref. 12. To begin, we define $\boldsymbol{\Gamma}(\theta) = \theta \boldsymbol{\mathcal{T}} + \boldsymbol{\mathcal{A}}$. Then,

$$\begin{aligned} -\ln\left(\frac{\det \boldsymbol{\mathcal{A}}}{\det(\boldsymbol{\mathcal{T}} + \boldsymbol{\mathcal{A}})} \right) &= \ln \det \boldsymbol{\Gamma}(\theta = 1) - \ln \det \boldsymbol{\Gamma}(\theta = 0) \\ &= \int d\mathbf{k} \int_{\boldsymbol{\Gamma}(\theta=0)}^{\boldsymbol{\Gamma}(\theta=1)} \frac{\delta \ln \det \boldsymbol{\Gamma}(\theta)}{\delta \widetilde{\boldsymbol{\Gamma}}^{-1}(\mathbf{k};\theta)} : \delta \widetilde{\boldsymbol{\Gamma}}^{-1}(\mathbf{k};\theta) \end{aligned} \tag{S16}$$

where the last equality is due to the chain rule, and the : symbol indicates scalar product of two tensors. Since $\det \boldsymbol{\Gamma}(\theta)$ can be expressed as a gaussian functional integral as in Eq. (S15), we have

$$\begin{aligned} &\frac{\delta \ln \det \boldsymbol{\Gamma}(\theta)}{\delta \widetilde{\boldsymbol{\Gamma}}^{-1}(\mathbf{k};\theta)} \\ &= \frac{\frac{1}{(2\pi)^3} \int \mathcal{D}\boldsymbol{\xi} \, \widetilde{\boldsymbol{\xi}}(\mathbf{k}) \widetilde{\boldsymbol{\xi}}(-\mathbf{k}) e^{-\frac{1}{2} \int \frac{d\mathbf{k}'}{(2\pi)^3} \widetilde{\boldsymbol{\xi}}(\mathbf{k}') \widetilde{\boldsymbol{\Gamma}}^{-1}(\mathbf{k}';\theta) \widetilde{\boldsymbol{\xi}}(-\mathbf{k}')}}{\int \mathcal{D}\boldsymbol{\xi} \, e^{-\frac{1}{2} \int \frac{d\mathbf{k}'}{(2\pi)^3} \widetilde{\boldsymbol{\xi}}(\mathbf{k}') \widetilde{\boldsymbol{\Gamma}}^{-1}(\mathbf{k}';\theta) \widetilde{\boldsymbol{\xi}}(-\mathbf{k}')}} \\ &= -\widetilde{\boldsymbol{\Gamma}}(\mathbf{k},\theta) \delta(\mathbf{k} = 0) \\ &= -\widetilde{\boldsymbol{\Gamma}}(\mathbf{k},\theta) \frac{V}{(2\pi)^3} \end{aligned} \tag{S17}$$

where, we have used the following interpretation of $\delta(\mathbf{k} = 0)$ in the last equality[42]:

$$\delta(\mathbf{k} = 0) = \frac{1}{(2\pi)^3} \int d\mathbf{r} \, e^{i\mathbf{k}\cdot\mathbf{r}}\big|_{\mathbf{k}=0} = \frac{1}{(2\pi)^3} \int d\mathbf{r} \, 1 = \frac{V}{(2\pi)^3} \tag{S18}$$

Furthermore, an expression for $\boldsymbol{\Gamma}^{-1}$ can be derived using a similar method that is used in Eq. (S29):

$$\widetilde{\boldsymbol{\Gamma}}^{-1}(\mathbf{k};\theta) = \frac{1}{\tilde{a}(\mathbf{k})} \left[\mathbb{1} - \frac{\varepsilon_0 \theta}{\beta\theta + \varepsilon_0 \tilde{a}(\mathbf{k})} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \right] \tag{S19}$$

Then, substituting Eqs. (S17) and (S19) into (S16), we have

$$\begin{aligned} &-\ln\left(\frac{\det \boldsymbol{\mathcal{A}}}{\det(\boldsymbol{\mathcal{T}} + \boldsymbol{\mathcal{A}})} \right) \\ &= \int \frac{d\mathbf{k}}{(2\pi)^3} \int_{\theta=0}^{1} \widetilde{\boldsymbol{\Gamma}}(\mathbf{k};\theta) : \frac{d\widetilde{\boldsymbol{\Gamma}}^{-1}(\mathbf{k};\theta)}{d\theta} d\theta \\ &= \int \frac{d\mathbf{k}}{(2\pi)^3} \int_{\theta=0}^{1} \left[\theta \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) + \widetilde{A}(\mathbf{k}) \right] : \frac{\varepsilon_0^2}{(\theta\beta + \varepsilon_0 \tilde{a}(\mathbf{k}))^2} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) d\theta \\ &= V \int \frac{d\mathbf{k}}{(2\pi)^3} \frac{\varepsilon_0}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \mathrm{tr} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) - V \int \frac{d\mathbf{k}}{(2\pi)^3} \left[\ln\left(\frac{\varepsilon_0 \tilde{a}(\mathbf{k})}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \right) + \frac{\beta}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \right] \\ &= -V \int \frac{d\mathbf{k}}{(2\pi)^3} \ln\left(\frac{\varepsilon_0 \tilde{a}(\mathbf{k})}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \right) \end{aligned} \tag{S20}$$

where last equality is due to $\mathrm{tr}\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) = \beta/\varepsilon_0$. Substituting Eq. (S20) into Eq. (S14), we have

$$\ln \Xi_0 = \frac{1}{2} V \int \frac{d\mathbf{k}}{(2\pi)^3} \ln\left(\frac{\varepsilon_0 \tilde{a}(\mathbf{k})}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})}\right) + \frac{1}{2}\int d\mathbf{r} \int d\mathbf{r}' \left[\boldsymbol{\mathcal{E}}_0(\mathbf{r}) - \boldsymbol{\mathcal{F}}(\mathbf{r})\right] \left[\boldsymbol{\mathcal{T}}(\mathbf{r}-\mathbf{r}') + \boldsymbol{\mathcal{A}}(\mathbf{r}-\mathbf{r}')\right]^{-1} \left[\boldsymbol{\mathcal{E}}_0(\mathbf{r}') - \boldsymbol{\mathcal{F}}(\mathbf{r}')\right] \quad \text{(S21)}$$

Next, we evaluate $\langle L - L_0 \rangle_0$, which can be further expressed as

$$\begin{aligned} &\langle L - L_0 \rangle_0 \\ &= -\sum_S \lambda_S \int d\mathbf{r} \int d\Omega_S \left\langle e^{i\int d\mathbf{r}' h_S(\mathbf{r}'-\mathbf{r})\boldsymbol{\mu}_S \cdot \boldsymbol{\mathcal{G}}(\mathbf{r}')} \right\rangle_0 - \frac{1}{2}\int d\mathbf{r} \int d\mathbf{r}' \langle [\boldsymbol{\mathcal{G}}(\mathbf{r}) + i\boldsymbol{\mathcal{F}}(\mathbf{r})] \boldsymbol{\mathcal{A}}^{-1}(\mathbf{r}-\mathbf{r}') [\boldsymbol{\mathcal{G}}(\mathbf{r}') + i\boldsymbol{\mathcal{F}}(\mathbf{r}')] \rangle_0 \end{aligned} \quad \text{(S22)}$$

The quantity $\left\langle e^{i\int d\mathbf{r}' h_S(\mathbf{r}'-\mathbf{r})\boldsymbol{\mu}_S \cdot \boldsymbol{\mathcal{G}}(\mathbf{r}')} \right\rangle_0$ can be evaluated using standard methods for gaussian integrals. The result is

$$\left\langle e^{i\int d\mathbf{r}' h_S(\mathbf{r}'-\mathbf{r})\boldsymbol{\mu}_S \cdot \boldsymbol{\mathcal{G}}(\mathbf{r}')} \right\rangle_0 = e^{-\frac{1}{2}\boldsymbol{\mu}_S \cdot \boldsymbol{\mathcal{T}}_{R,S} \cdot \boldsymbol{\mu}_S + \boldsymbol{\gamma}_S(\mathbf{r}) \cdot \boldsymbol{\mu}_S} \quad \text{(S23)}$$

where

$$\boldsymbol{\mathcal{T}}_{R,S} = \int \frac{d\mathbf{k}}{(2\pi)^3} \frac{\varepsilon_0 \tilde{a}(\mathbf{k}) \tilde{h}_S(\mathbf{k})^2}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \quad \text{(S24)}$$

and

$$\widetilde{\boldsymbol{\gamma}}_S(\mathbf{k}) = \tilde{h}_S(\mathbf{k}) \left[\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) - \frac{\varepsilon_0}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \left(\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k})\right)\right] \quad \text{(S25)}$$

The quantity $\langle [\boldsymbol{\mathcal{G}}(\mathbf{r}) + i\boldsymbol{\mathcal{F}}(\mathbf{r})] \boldsymbol{\mathcal{A}}^{-1}(\mathbf{r}-\mathbf{r}') [\boldsymbol{\mathcal{G}}(\mathbf{r}') + i\boldsymbol{\mathcal{F}}(\mathbf{r}')] \rangle_0$ can also be evaluated using standard techniques for gaussian integrals. Letting $\boldsymbol{\xi}(\mathbf{r}) = \boldsymbol{\mathcal{G}}(\mathbf{r}) + i\boldsymbol{\mathcal{F}}(\mathbf{r})$, we can write

$$\begin{aligned} &\left\langle \boldsymbol{\xi}(\mathbf{r}) \boldsymbol{\mathcal{A}}^{-1}(\mathbf{r}-\mathbf{r}') \boldsymbol{\xi}(\mathbf{r}') \right\rangle_0 \\ &= \frac{1}{\Xi_0} \int \mathcal{D}\mathbf{P} \int \mathcal{D}\boldsymbol{\mathcal{G}}\, \boldsymbol{\xi}(\mathbf{r}) \boldsymbol{\mathcal{A}}^{-1}(\mathbf{r}-\mathbf{r}') \boldsymbol{\xi}(\mathbf{r}') e^{-L_0[\mathbf{P},\boldsymbol{\mathcal{G}}]} \\ &= \frac{1}{\Xi_0} \frac{\delta}{i\delta \mathbf{J}(\mathbf{r})} \cdot \boldsymbol{\mathcal{A}}^{-1}(\mathbf{r}-\mathbf{r}') \cdot \frac{\partial}{i\partial \mathbf{J}(\mathbf{r}')} \left(\int \mathcal{D}\mathbf{P} \int \mathcal{D}\boldsymbol{\xi}\, e^{-L_0[\mathbf{P},\boldsymbol{\mathcal{G}}] - i\int d\mathbf{r}_1 \mathbf{J}(\mathbf{r}_1)\cdot\boldsymbol{\xi}(\mathbf{r}_1)} \right)\Big|_{\mathbf{J}=\mathbf{0}} \end{aligned} \quad \text{(S26)}$$

Evaluation of the above integral gives

$$\begin{aligned} &\int d\mathbf{r} \int d\mathbf{r}' \left\langle \boldsymbol{\xi}(\mathbf{r}) \boldsymbol{\mathcal{A}}^{-1}(\mathbf{r}-\mathbf{r}') \boldsymbol{\xi}(\mathbf{r}') \right\rangle_0 \\ &= \int \frac{d\mathbf{k}}{(2\pi)^3} V \frac{\beta}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} - \int \frac{d\mathbf{k}}{(2\pi)^3} \frac{1}{\tilde{a}(\mathbf{k})} \left[\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k})\right] \left(\mathbb{1} - \frac{\varepsilon_0}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\right)^2 \left[\widetilde{\boldsymbol{\mathcal{E}}}_0(-\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(-\mathbf{k})\right] \end{aligned} \quad \text{(S27)}$$

In the evaluation of Eqs. (S21), (S23), and (S27), we have used the following relation:

$$[\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) + \widetilde{\boldsymbol{\mathcal{A}}}(\mathbf{k})]^{-1} = \frac{1}{\tilde{a}(\mathbf{k})} \left[\mathbb{1} - \frac{\varepsilon_0}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\right] \quad \text{(S28)}$$

The evaluation of the above expression uses the following strategy:

$$\begin{aligned} &[\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) + \widetilde{\boldsymbol{\mathcal{A}}}(\mathbf{k})]^{-1} \\ &= [\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) + \tilde{a}(\mathbf{k})\mathbb{1}]^{-1} \\ &= \frac{1}{\tilde{a}(\mathbf{k})} [\mathbb{1} + \tilde{a}(\mathbf{k})^{-1} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})]^{-1} \\ &= \frac{1}{\tilde{a}(\mathbf{k})} [\mathbb{1} - \tilde{a}(\mathbf{k})^{-1} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) + \tilde{a}(\mathbf{k})^{-2} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})^2 - \tilde{a}(\mathbf{k})^{-3} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})^3 + \ldots] \\ &= \frac{1}{\tilde{a}(\mathbf{k})} \left[\mathbb{1} - \left(\frac{\beta}{\varepsilon_0 \tilde{a}(\mathbf{k})}\right) \frac{\mathbf{k}\mathbf{k}}{k^2} + \left(\frac{\beta}{\varepsilon_0 \tilde{a}(\mathbf{k})}\right)^2 \frac{\mathbf{k}\mathbf{k}}{k^2} - \left(\frac{\beta}{\varepsilon_0 \tilde{a}(\mathbf{k})}\right)^3 \frac{\mathbf{k}\mathbf{k}}{k^2} + \ldots\right] \\ &= \frac{1}{\tilde{a}(\mathbf{k})} \left[\mathbb{1} - \frac{\beta}{\varepsilon_0 \tilde{a}(\mathbf{k})} \frac{1}{1 + \frac{\beta}{\varepsilon_0 \tilde{a}(\mathbf{k})}} \frac{\mathbf{k}\mathbf{k}}{k^2}\right] \\ &= \frac{1}{\tilde{a}(\mathbf{k})} \left[\mathbb{1} - \frac{\varepsilon_0}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\right] \end{aligned} \quad \text{(S29)}$$

In the fourth equality above, we have used the fact that $\mathbf{kk}/k^2$ is a projection operator, and thus, $(\mathbf{kk}/k^2)^n = \mathbf{kk}/k^2$ for any positive integer n.

D. Stationary value of the variational grand potential

We approximate the free energy of the mixture by taking the stationary value of W with respect to the variational parameters a and $\boldsymbol{\mathcal{F}}$. As such, the values of a and $\boldsymbol{\mathcal{F}}$ are set by solving the following equations:

$$\frac{\partial W}{\partial \tilde{a}(\mathbf{k})} = 0 \tag{S30}$$

and

$$\frac{\partial W}{\partial \widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k})} = 0 \tag{S31}$$

Eqs. (S30) and (S31) lead to

$$\begin{aligned}
&\frac{1}{2}V\frac{\beta^2}{\tilde{a}(\mathbf{k})\left[\beta+\varepsilon_0\tilde{a}(\mathbf{k})\right]^2} - \left(\frac{\varepsilon_0}{\beta+\varepsilon_0\tilde{a}(\mathbf{k})}\right)^3 \left[\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k})\right] \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \left[\widetilde{\boldsymbol{\mathcal{E}}}_0(-\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(-\mathbf{k})\right] \\
&= \sum_S \lambda_s \int d\mathbf{r} \int d\Omega_s \\
&\qquad \left[\frac{\beta\varepsilon_0\tilde{h}_s(\mathbf{k})^2}{2(\beta+\varepsilon_0\tilde{a}(\mathbf{k}))^2}\boldsymbol{\mu}_s\cdot\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\cdot\boldsymbol{\mu}_s - \tilde{h}_s(\mathbf{k})\left(\frac{\varepsilon_0}{\beta+\varepsilon_0\tilde{a}(\mathbf{k})}\right)^2 \boldsymbol{\mu}_s\cdot\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\cdot[\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k})]e^{i\mathbf{k}\cdot\mathbf{r}}\right] e^{-f_S(\boldsymbol{\mu}_S,\mathbf{r})}
\end{aligned} \tag{S32}$$

and

$$\frac{1}{\tilde{h}_s(\mathbf{k})}\left(\frac{\varepsilon_0}{\beta+\varepsilon_0\tilde{a}(\mathbf{k})}\right)\left[\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k})\right] = \sum_S \lambda_s \int d\mathbf{r} \int d\Omega_s\, \boldsymbol{\mu}_s e^{-f_S(\boldsymbol{\mu}_S,\mathbf{r}) - i\mathbf{k}\cdot\mathbf{r}} \tag{S33}$$

Additionally, the fugacity λ_s can be related to the fluid density ρ_s through

$$\rho_s V = -\frac{\partial W}{\partial \mu_s} = \lambda_s \int d\mathbf{r} \int d\Omega_s\, e^{-f_S(\boldsymbol{\mu}_S,\mathbf{r})} \tag{S34}$$

E. Simplification with point-dipole approximation whenever applicable

When the point-dipole limit $h_s(\mathbf{r}) = \delta(\mathbf{r})$ does not cause numerical divergences, the short-wavelength nature of the molecular polarization is not physically important. Therefore, we simplify the theory by taking the limit $h_s(\mathbf{r}) = \delta(\mathbf{r})$, i.e. $\tilde{h}_s(\mathbf{k}) = 1$, whenever this procedure does not lead to numerical divergences. With this approximation, the resulting theory is given by

$$\begin{aligned}
\beta W = &-\frac{1}{2}\int \frac{d\mathbf{k}}{(2\pi)^3} V \left[\ln\left(\frac{\varepsilon_0\tilde{a}(\mathbf{k})}{\beta+\varepsilon_0\tilde{a}(\mathbf{k})}\right) + \frac{\beta}{\beta+\varepsilon_0\tilde{a}(\mathbf{k})}\right] \\
&-\frac{1}{2}\int \frac{d\mathbf{k}}{(2\pi)^3} \left(\frac{\varepsilon_0}{\beta+\varepsilon_0\tilde{a}(\mathbf{k})}\right)^2 \left[\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k})\right] \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \left[\widetilde{\boldsymbol{\mathcal{E}}}_0(-\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(-\mathbf{k})\right] \\
&-\sum_S \lambda_s \int d\mathbf{r} \int d\Omega_s\, e^{-f_S(\boldsymbol{\mu}_S,\mathbf{r})}
\end{aligned} \tag{S35}$$

where the effective single-molecule potential is

$$f_s(\boldsymbol{\mu}_S,\mathbf{r}) = \frac{1}{2}\,\boldsymbol{\mu_s}\cdot\boldsymbol{\mathcal{T}}_{R,S}\cdot\boldsymbol{\mu}_S - \boldsymbol{\mu}_S\cdot\boldsymbol{\gamma}_S(\mathbf{r}) \tag{S36}$$

with

$$\boldsymbol{\mathcal{T}}_{R,S} = \int \frac{d\mathbf{k}}{(2\pi)^3} \frac{\varepsilon_0\tilde{a}(\mathbf{k})\tilde{h}_S(\mathbf{k})^2}{\beta+\varepsilon_0\tilde{a}(\mathbf{k})} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \tag{S37}$$

and

$$\widetilde{\boldsymbol{\gamma}}_s(\mathbf{k}) = \widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) - \frac{\varepsilon_0}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \left(\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k}) \right) \tag{S38}$$

Furthermore, the stationary values of the variational parameters are given by the solutions to the following equations

$$\frac{\beta V}{\varepsilon_0 \tilde{a}(\mathbf{k})} = \sum_S \lambda_s \int d\mathbf{r} \int d\Omega_s \, \boldsymbol{\mu}_s \cdot \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \cdot \boldsymbol{\mu}_s e^{-f_S(\boldsymbol{\mu}_S, \mathbf{r})} \tag{S39}$$

and

$$\left(\frac{\varepsilon_0}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \right) \left[\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) - \widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k}) \right] = \sum_S \lambda_s \int d\mathbf{r} \int d\Omega_s \, \boldsymbol{\mu}_s e^{-f_S(\boldsymbol{\mu}_S, \mathbf{r}) - i\mathbf{k}\cdot\mathbf{r}} \tag{S40}$$

F. The variational parameters in the linear response regime

We now solve Eqs. (S39) and (S40) in the linear response regime, i.e. the situation when the applied field is weak. In this regime, the polarization of the fluid responds to the applied field linearly, and therefore, we may solve Eqs. (S39) and (S40) to first order in the applied field. At this order, Eq. (S39) reduces to

$$\begin{aligned} &\frac{\beta V}{\varepsilon_0 \tilde{a}(\mathbf{k})} \\ &= \sum_S \lambda_s \int d\mathbf{r} \int d\Omega_s \, \boldsymbol{\mu}_s \cdot \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \cdot \boldsymbol{\mu}_s \, e^{-\frac{1}{2}\boldsymbol{\mu}_S \cdot \boldsymbol{\mathcal{T}}_R \cdot \boldsymbol{\mu}_S} \left(1 + \boldsymbol{\gamma}_s(\mathbf{r}) \cdot \boldsymbol{\mu}_s\right) \\ &= \sum_S \lambda_s \int d\mathbf{r} \int d\Omega_s \, \boldsymbol{\mu}_s \cdot \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \cdot \boldsymbol{\mu}_s \, e^{-\frac{1}{2}\boldsymbol{\mu}_S \cdot \boldsymbol{\mathcal{T}}_R \cdot \boldsymbol{\mu}_S} \end{aligned} \tag{S41}$$

where the second equality results from the integral over an odd power of the permanent dipole moment. Eq. (S41) suggests that $\tilde{a}(\mathbf{k})$ is isotropic with respect to $\mathbf{k}$ in the linear response regime. Consequently, the effective self-interaction tensor $\boldsymbol{\mathcal{T}}_{R,S}$ is also isotropic and we may write

$$\boldsymbol{\mathcal{T}}_{R,S} = \int \frac{d\mathbf{k}}{(2\pi)^3} \frac{\varepsilon_0 \tilde{a}(\mathbf{k})}{\beta + \varepsilon_0 \tilde{a}(\mathbf{k})} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) = T_{R,S} \mathbb{1} \tag{S42}$$

where $T_{R,S}$ characterizes the strength of the effective self-interaction for a molecule of species S. Because $\boldsymbol{\mathcal{T}}_{R,S}$ is isotropic, the term $\frac{1}{2}\boldsymbol{\mu}_S \cdot \boldsymbol{\mathcal{T}}_{R,S} \cdot \boldsymbol{\mu}_S = \frac{1}{2} T_{R,S} \bar{\mu}_S^2$ is independent of the molecule's spatial position and the dipole moment direction. With the simplification given by Eq. (S42), the relation between fluid density and the fugacity reduces to

$$\rho_S = 4\pi \lambda_S e^{-\frac{1}{2} T_{R,S} \bar{\mu}_S^2} \tag{S43}$$

Eq. (S41) simplifies to the following, giving the solution to $\tilde{a}(\mathbf{k})$ in the linear response regime:

$$\frac{\beta}{\varepsilon_0 \tilde{a}(\mathbf{k})} = \sum_S \frac{\beta \rho_S \bar{\mu}_S^2}{3\varepsilon_0} = y \tag{S44}$$

where we have defined a dimensionless parameter $y = \sum_S \frac{\beta \rho_S \bar{\mu}_S^2}{3\varepsilon_0}$ that characterizes the strength of dipolar interactions in the liquid. On the other hand, Eq. (S40) simplifies to

$$\left(\mathbb{1} + y \frac{\varepsilon_0}{\beta} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \right) \widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k}) = -y \left(\mathbb{1} - \frac{\varepsilon_0}{\beta} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \right) \widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) \tag{S45}$$

For $\mathbf{k} \neq \mathbf{0}$, the matrix $\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})$ is well defined, and therefore, we can express $\widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k})$ as

$$\widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k}) = -y \left(\mathbb{1} + y \frac{\varepsilon_0}{\beta} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \right)^{-1} \left(\mathbb{1} - \frac{\varepsilon_0}{\beta} \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \right) \widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) \tag{S46}$$

The inverse operator of $\mathbb{1} + y\frac{\varepsilon_0}{\beta}\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})$ can be calculated using a similar strategy as that presented in Eq. (S29), which gives

$$\left(\mathbb{1} + y\frac{\varepsilon_0}{\beta}\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\right)^{-1} = \mathbb{1} - \frac{y}{1+y}\frac{\varepsilon_0}{\beta}\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \tag{S47}$$

And therefore, for $\mathbf{k} \neq \mathbf{0}$:

$$\widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k}) = -y\left(\mathbb{1} - \frac{\varepsilon_0}{\beta}\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\right)\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) \qquad \text{for } \mathbf{k} \neq \mathbf{0} \tag{S48}$$

For $\mathbf{k} = 0$, $\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})$ is not well-defined in the $\mathbf{k}$-space. The expression in Eq. (S45) is best solved in the position space instead. We obtain the equation in the position space by inverse Fourier transform of Eq. (S45), which gives

$$\int d\mathbf{r}' \left(\mathbb{1}\delta(\mathbf{r}-\mathbf{r}') + y\frac{\varepsilon_0}{\beta}\boldsymbol{\mathcal{T}}(\mathbf{r}-\mathbf{r}')\right)\boldsymbol{\mathcal{F}}(\mathbf{r}') = -\int d\mathbf{r}'\, y\left(\mathbb{1}\delta(\mathbf{r}-\mathbf{r}') - \frac{\varepsilon_0}{\beta}\boldsymbol{\mathcal{T}}(\mathbf{r}-\mathbf{r}')\right)\boldsymbol{\mathcal{E}}_0(\mathbf{r}') \tag{S49}$$

The $\mathbf{k} = \mathbf{0}$ values of $\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k})$ and $\widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k})$ contribute to spatially uniform fields in the $\mathbf{r}$ space. For uniform $\boldsymbol{\mathcal{E}}_0(\mathbf{r})$ and $\boldsymbol{\mathcal{F}}(\mathbf{r})$, Eq. (S49) becomes

$$\int d\mathbf{r}' \left(\mathbb{1}\delta(\mathbf{r}-\mathbf{r}') + y\frac{\varepsilon_0}{\beta}\boldsymbol{\mathcal{T}}(\mathbf{r}-\mathbf{r}')\right)\boldsymbol{\mathcal{F}} = -\int d\mathbf{r}'\, y\left(\mathbb{1}\delta(\mathbf{r}-\mathbf{r}') - \frac{\varepsilon_0}{\beta}\boldsymbol{\mathcal{T}}(\mathbf{r}-\mathbf{r}')\right)\boldsymbol{\mathcal{E}}_0 \tag{S50}$$

which then gives

$$\left(1 + \frac{y}{3}\right)\boldsymbol{\mathcal{F}} = -y\left(1 - \frac{1}{3}\right)\boldsymbol{\mathcal{E}}_0 \tag{S51}$$

based on the expression for $\mathbf{T}(\mathbf{r})$ in Eq. (S3). Therefore, for $\mathbf{k} = \mathbf{0}$:

$$\widetilde{\boldsymbol{\mathcal{F}}}(\mathbf{k}) = -\frac{2y}{y+3}\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) \qquad \text{for } \mathbf{k} = \mathbf{0} \tag{S52}$$

Additionally, the grand potential simplifies to the following expression in the linear response regime:

$$\begin{aligned}\beta W = &-\frac{1}{2}\int \frac{d\mathbf{k}}{(2\pi)^3} V\left[\frac{y}{y+1} - \ln(1+y)\right] \\ &-\frac{1}{2}\int \frac{d\mathbf{k}}{(2\pi)^3}\left(\frac{\varepsilon_0}{\beta}\right)^2\left(\frac{y}{1+y}\right)^2 \widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k})\left[\mathbb{1} - \widetilde{\mathbf{K}}(\mathbf{k})\right]\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\left[\mathbb{1} - \widetilde{\mathbf{K}}(-\mathbf{k})\right]\widetilde{\boldsymbol{\mathcal{E}}}_0(-\mathbf{k}) \\ &-\sum_S \rho_s V - \sum_S \frac{\rho_s \bar{\mu}_s^2}{6}\int \frac{d\mathbf{k}}{(2\pi)^3}\boldsymbol{\gamma}_s(\mathbf{k})\cdot\boldsymbol{\gamma}_s(-\mathbf{k})\end{aligned} \tag{S53}$$

G. The polarization and the dielectric constant

We compute the dielectric constant of the mixture by considering the variation in the polarization with the applied field. To derive the polarization-applied field relation, we take the derivative of the grand potential with respect to the applied field:

$$\mathbf{P}(\mathbf{r}) = -\frac{\delta\beta W}{\delta\boldsymbol{\mathcal{E}}_0(\mathbf{r})} \tag{S54}$$

In the $\mathbf{k}$-space, this can be equivalently written as

$$\widetilde{\mathbf{P}}(\mathbf{k}) = -(2\pi)^3\frac{\delta\beta W}{\delta\widetilde{\boldsymbol{\mathcal{E}}}_0(-\mathbf{k})} \tag{S55}$$

Performing the derivative leads to

$$\widetilde{\mathbf{P}}(\mathbf{k}) = \left(\frac{\varepsilon_0}{\beta + \varepsilon_0\tilde{a}}\right)^2\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\left[\mathbb{1} - \widetilde{\mathbf{K}}(\mathbf{k})\right]^2\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) + \frac{1}{\tilde{a}}\left\{\mathbb{1} - \left(\frac{\varepsilon_0}{\beta + \varepsilon_0\tilde{a}}\right)\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\left[\mathbb{1} - \widetilde{\mathbf{K}}(\mathbf{k})\right]\right\}^2\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{k}) \tag{S56}$$

The electric susceptibility $\widetilde{\boldsymbol{\chi}}_0$ of the system relates the polarization of the mixture to the applied electric field through $\widetilde{\mathbf{P}}(\mathbf{k}) = \varepsilon_0 \widetilde{\boldsymbol{\chi}}_0(\mathbf{k}) \widetilde{\mathbf{E}}_0(\mathbf{k})$. From Eq. (S56), we obtain the expression for $\widetilde{\boldsymbol{\chi}}_0$, given by

$$\widetilde{\boldsymbol{\chi}}_0(\mathbf{k}) = \frac{\beta}{\varepsilon_0}\left(\frac{\varepsilon_0}{\beta+\varepsilon_0\tilde{a}}\right)^2 \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\left[\mathbb{1} - \widetilde{\mathbf{K}}(\mathbf{k})\right]^2 + \frac{\beta}{\varepsilon_0\tilde{a}}\left\{\mathbb{1} - \left(\frac{\varepsilon_0}{\beta+\varepsilon_0\tilde{a}}\right)\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\left[\mathbb{1} - \widetilde{\mathbf{K}}(\mathbf{k})\right]\right\}^2 \tag{S57}$$

which can be simplified to

$$\widetilde{\boldsymbol{\chi}}_0(\mathbf{k}) = y\mathbb{1} - \left(\frac{y^2}{y+1}\right)\frac{\varepsilon_0}{\beta}\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})\left[\mathbb{1} - \widetilde{\mathbf{K}}(\mathbf{k})^2\right] \tag{S58}$$

At $\mathbf{k} = 0$, we have

$$\widetilde{\boldsymbol{\chi}}_0(\mathbf{k}=\mathbf{0}) = y\mathbb{1} - \left(\frac{y^2}{y+1}\right)\frac{\varepsilon_0}{\beta}\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}=\mathbf{0})\left[\mathbb{1} - \left(\frac{2y}{y+3}\right)^2\right] \tag{S59}$$

To obtain the dielectric constant of the mixture, we use the relation[35]

$$\frac{(\varepsilon-1)(2\varepsilon+1)}{\varepsilon} = \mathrm{tr}\,\widetilde{\boldsymbol{\chi}}_0(\mathbf{k}=\mathbf{0}) \tag{S60}$$

Noting that $\mathrm{tr}\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}=\mathbf{0}) = \beta/\varepsilon_0$, we arrive at the following simple expression for dielectric constant of the mixture:

$$\frac{(\varepsilon-1)(2\varepsilon+1)}{\varepsilon} = 3y\left(\frac{2y^2+3y+9}{(y+3)^2}\right) \tag{S61}$$

H. A consistent interpretation of the momentum cut-off

Due to the infinite upper limit in the momentum integral, the grand potential in Eq. (S53) is divergent as written. A momentum cut-off is needed. We write the cutoff as $2\pi/b$, where b is a microscopic length scale on the order of the molecular size. With the cutoff, we can write

$$\int \frac{d\mathbf{k}}{(2\pi)^3} 1 = \frac{1}{b^3} \tag{S62}$$

The value of b is usually taken as the size of a molecule, the lattice spacing, or be treated as a fitting parameter. However, in this work, we have a consistent way of defining the value of b in the linear response regime, relating it to the short-range function $h_s(\mathbf{r})$ that defines the length scale of the molecular polarization. Starting from Eq. (S32), we consider the situation where $\boldsymbol{\mathcal{E}}_0 = \mathbf{0}$, which gives

$$V\frac{\beta}{\beta+\varepsilon_0\tilde{a}(\mathbf{k})} = \sum_S \lambda_s \int d\mathbf{r} \int d\Omega_s \left[\frac{\varepsilon_0\tilde{a}(\mathbf{k})\tilde{h}_s(\mathbf{k})^2}{(\beta+\varepsilon_0\tilde{a}(\mathbf{k}))}\boldsymbol{\mu}_s \cdot \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \cdot \boldsymbol{\mu}_s\right] e^{-\frac{1}{2}T_{R,S}\bar{\mu}_S^2} \tag{S63}$$

The above expression can be further simplified with the help of Eq. (S43), giving

$$V\frac{\beta}{\beta+\varepsilon_0\tilde{a}(\mathbf{k})} = \sum_S \frac{\rho_s}{4\pi} \int d\mathbf{r} \int d\Omega_s \left[\frac{\varepsilon_0\tilde{a}(\mathbf{k})\tilde{h}_s(\mathbf{k})^2}{(\beta+\varepsilon_0\tilde{a}(\mathbf{k}))}\boldsymbol{\mu}_s \cdot \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \cdot \boldsymbol{\mu}_s\right] \tag{S64}$$

Integrating the both sides of Eq. (S64) over the momentum space, we have

$$\begin{aligned}\int \frac{d\mathbf{k}}{(2\pi)^3} V\frac{\beta}{\beta+\varepsilon_0\tilde{a}(\mathbf{k})} &= \int \frac{d\mathbf{k}}{(2\pi)^3} \sum_S \frac{\rho_s}{4\pi} \int d\mathbf{r} \int d\Omega_s \left[\frac{\varepsilon_0\tilde{a}(\mathbf{k})\tilde{h}_s(\mathbf{k})^2}{(\beta+\varepsilon_0\tilde{a}(\mathbf{k}))}\boldsymbol{\mu}_s \cdot \widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \cdot \boldsymbol{\mu}_s\right] \\ &= \sum_S \rho_s V T_{R,S}\bar{\mu}_S^2 \end{aligned} \tag{S65}$$

where the second equality is due to Eqs. (S37) and (S42). Noting that $\tilde{a}$ is independent of $\mathbf{k}$ in the linear response regime, we integrate the left-hand-side of Eq. (S65) to the momentum cut-off value, arriving at

$$\frac{1}{b^3} = \frac{\beta+\varepsilon_0\tilde{a}}{\beta}\sum_S \rho_s T_{R,S}\bar{\mu}_S^2 \tag{S66}$$

Eq. (S66) provides us a consistent interpretation of the momentum cutoff. Note that the effect of the cut-off resides in $T_{R,S}$, which describes the effective self-interaction. In the linear response regime, we have

$$T_{R,S}\mathbb{1} = \frac{\varepsilon_0\tilde{a}}{\beta+\varepsilon_0\tilde{a}}\int\frac{d\mathbf{k}}{(2\pi)^3}\tilde{h}_S(\mathbf{k})^2\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) \tag{S67}$$

We note that the momentum integral of the dipole-dipole interaction tensor $\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})$ is regularized by the function h_S in the above expression for $T_{R,S}$.

I. The choice of the molecular polarization distribution function

In this section, we look into how the molecular polarization distribution function h_S regularizes the dipole-dipole interaction tensor. In particular, we consider the integral $\int\frac{d\mathbf{k}}{(2\pi)^3}\tilde{h}_S(\mathbf{k})^2\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})$ that appears in the computation of the effective self-interaction tensor $\boldsymbol{\mathcal{T}}_{R,S}$.

Let us first consider the following function h_S that describes a uniform distribution of the polarization within a sphere of volume v_S', where v_S' is the volume of the molecular dipole moment of species S. That is

$$h_S(\mathbf{r}-\mathbf{r}_{S,i}) = \begin{cases}\frac{1}{v_S'} & \text{if } |\mathbf{r}-\mathbf{r}_{S,i}| < \left(\frac{3v_S'}{4\pi}\right)^{\frac{1}{3}} \\ 0 & \text{otherwise}\end{cases} \tag{S68}$$

To compute the integral $\int\frac{d\mathbf{k}}{(2\pi)^3}\tilde{h}_S(\mathbf{k})^2\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k})$ for this function, we first note that this integral can be rewritten as position-space integral as

$$\int\frac{d\mathbf{k}}{(2\pi)^3}\tilde{h}_S(\mathbf{k})^2\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) = \int d\mathbf{r}_1\int d\mathbf{r}_2\, h_S(\mathbf{r}-\mathbf{r}_1)\boldsymbol{\mathcal{T}}(\mathbf{r}_1-\mathbf{r}_2)h_S(\mathbf{r}_2-\mathbf{r}) \tag{S69}$$

The right-hand-side of Eq. (S69) describes the energy where a uniformly polarized sphere that has a unit dipole moment interacts with the electric field generated by itself. This problem has been solved in many textbooks of electrostatics, for instance, in Example 4.2 in Ref. 43. Knowing that the electric field inside a uniformly polarized sphere or polarization $\mathbf{P}$ is $-\mathbf{P}/(3\varepsilon_0)$, we can write down

$$\int\frac{d\mathbf{k}}{(2\pi)^3}\tilde{h}_S(\mathbf{k})^2\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) = \frac{\beta}{3\varepsilon_0 v_S'}\mathbb{1} \tag{S70}$$

Let us also consider an alternative form of the function h_S that describes the molecular distribution as a Gaussian function of width σ_S, i.e.

$$h_S(\mathbf{r}) = \frac{1}{(2\pi\sigma_S^2)^{\frac{3}{2}}}\exp\left(-\frac{\mathbf{r}^2}{2\sigma_S^2}\right) \tag{S71}$$

The Fourier transform of $h_S(\mathbf{r})$ is

$$\tilde{h}_S(\mathbf{k}) = \exp\left(-\frac{\sigma_S^2\mathbf{k}^2}{2}\right) \tag{S72}$$

Then, the following integral can be directly evaluated as

$$\int\frac{d\mathbf{k}}{(2\pi)^3}\tilde{h}_S(\mathbf{k})^2\widetilde{\boldsymbol{\mathcal{T}}}(\mathbf{k}) = \frac{\beta}{3\varepsilon_0(2\sqrt{\pi}\sigma_S)^3}\mathbb{1} \tag{S73}$$

Comparing Eqs. (S70) and (S73), we note that the two functions of $h_S(\mathbf{r})$ give the same results if we set $v_S' = (2\sqrt{\pi}\sigma_S)^3$. In general, the values of v_S' and σ_S may be treated as an adjustable parameter to be obtained by fitting the experimental data of the dielectric constant of the pure components. However, in order to make *a priori* predictions free of fitting parameters, we connect it to the molecular volume by $v_S = v_S' = (2\sqrt{\pi}\sigma_S)^3$, with v_S being the molecular volume. Therefore,

$$\int\frac{d\mathbf{k}}{(2\pi)^3}1 = \frac{1}{b^3} = \frac{\varepsilon_0\tilde{a}}{\beta}\sum_S\frac{\beta\bar{\mu}_S^2\rho_S}{3\varepsilon_0 v_S} \tag{S74}$$

While we have chosen two spherically symmetrical functions for $h_S(\mathbf{r})$ in this section, we note that $h_S(\mathbf{r})$ can be more generally chosen to describe molecular polarization that is more complex. For example, the polarization distribution of a single molecule may be calculated with *ab initio* quantum-mechanical calculations. This may allow the theory to capture the specific dipole-dipole interactions due to the complex chemical structure of the molecules.

J. Free energies of a homogeneous mixture

It is of interest to consider the free energy of a homogeneous mixture in the linear response regime. In a uniform applied field $|\mathbf{E}_0| = E_0$, we have

$$\begin{aligned} |\boldsymbol{\mathcal{E}}_0| &= \mathcal{E}_0 = \beta E_0 \\ |\boldsymbol{\mathcal{F}}| &= -\frac{2y}{y+3}\mathcal{E}_0 \\ |\boldsymbol{\gamma}| &= \frac{3}{y+3}\mathcal{E}_0 \end{aligned} \tag{S75}$$

The expression for the grand potential for a homogeneous mixture is reduced to

$$\beta W = -\frac{1}{2}\left[\frac{y}{y+1} - \ln(y+1)\right]\sum_S \frac{y_S^{(m)} V}{y v_S} - \sum_S \rho_S V - \frac{1}{2}\beta V\left[\frac{3\left(y^2 + 3\sum_S y_S^{(m)}\right)}{(y+3)^2}\right]\varepsilon_0 E_0^2 \tag{S76}$$

where $y_S^{(m)} = \beta\bar{\mu}_S^2\rho_S/(3\varepsilon_0)$, with ρ_S being the density of species S inside the mixture.

The Helmholtz free energy F of the mixture can be derived by performing a Legendre transform on the grand potential through

$$F = W + \sum_S \mu_S \rho_S V \tag{S77}$$

The transformation gives

$$\beta F = \frac{1}{2}\ln(y+1)\sum_S \frac{y_S^{(m)} V}{y v_S} - \frac{1}{2}\beta V\left[\frac{3\left(y^2 + 3\sum_S y_S^{(m)}\right)}{(y+3)^2}\right]\varepsilon_0 E_0^2 - \sum_S\left[\rho_S V \ln\left(\rho_S \Lambda_S^3\right) - \rho_S V\right] \tag{S78}$$

The free energy of mixing, ΔF_{mix}, can be calculated by subtracting the free energy of the mixture from that of an unmixed system. Assuming that there is no volume change upon mixing, we can write ΔF_{mix} for a binary mixture as

$$\Delta F_{\text{mix}} = F(\phi_A, \phi_B) - \phi_A F(\phi_A = 1, \phi_B = 0) - \phi_B F(\phi_A = 0, \phi_B = 1) \tag{S79}$$

where ϕ_S is the volume fraction of species S in the mixture given by $\phi_S = \rho_S v_S$. In the absence of external fields, evaluation of Eq. (S79) leads to Eq. (9) in the main manuscript.

S2. CONSIDERATION OF LIQUID INCOMPRESSIBILITY

In this section, we introduce an incompressibility constraint for the liquid explicitly. We show that, at the self-consistent-field level, the resulting theory is the same as that presented in Sec. S1.

We consider a grand canonical ensemble of the fluid mixture under chemical potential μ'_S for each species S at temperature T and volume V. The grand partition function of the system is given by

$$\Xi' = \sum_{N_A=0}^{\infty} \cdots \sum_{N_B=0}^{\infty} \frac{e^{\beta\mu'_A N_A}}{N_A!} \cdots \frac{e^{\beta\mu'_B N_B}}{N_B!} Z'(\{N_S\}) \tag{S80}$$

In this section, we use primed quantities (such as Ξ' instead of Ξ) to indicate that they are different from those in Sec. S1. $Z'(\{N_S\})$ is the canonical partition function with an explicitly considered incompressibility condition, given by

$$Z'(\{N_S\}) = \prod_S \prod_{i=1}^{N_S} \int \frac{d\mathbf{r}_{S,i}}{\Lambda_S^3} \int \frac{d\Omega_{S,i}}{4\pi} \delta\left[\sum_S v_S \hat{\rho}_S(\mathbf{r}) - 1\right] e^{-\beta U} \tag{S81}$$

where the incompressibility condition of the liquid is enforced by a δ-functional. This condition considers the effects of the nonelectrostatic intermolecular forces and controls the density of the liquid at a given temperature and pressure. In the δ-functional, $\hat{\rho}_s(\mathbf{r}) = \sum_{i=1}^{N_S} \delta(\mathbf{r} - \mathbf{r}_{s,i})$ is the number density operator for solvent S, while v_s is the average volume of each molecule of species S.

By rewriting the δ-functional using its Fourier representation, we arrive at

$$\begin{aligned} Z'(\{N_S\}) &= \prod_S \prod_{i=1}^{N_S} \int \frac{d\mathbf{r}_{S,i}}{\Lambda_S^3} \int \frac{d\Omega_{S,i}}{4\pi} \int \mathcal{D}w\, e^{i\int d\mathbf{r} w(\mathbf{r})\left[\sum_S v_S \hat{\rho}_S(\mathbf{r}) - 1\right]} e^{-\beta U} \\ &= \prod_S \prod_{i=1}^{N_S} \int \frac{d\mathbf{r}_{S,i}}{\Lambda_S^3} \int \frac{d\Omega_{S,i}}{4\pi} \int \mathcal{D}w\, e^{\sum_S i v_S w(\mathbf{r}_{S,i}) - i\int d\mathbf{r} w(\mathbf{r})} e^{-\beta U} \end{aligned} \tag{S82}$$

Substituting $Z'\{N_s\})$ into the grand partition function Ξ', we write the field-based expression for Ξ' as:

$$\Xi' = \int \mathcal{D}\mathbf{P} \int \mathcal{D}\boldsymbol{\mathcal{G}} \int \mathcal{D}w\, e^{-L'[\mathbf{P},\boldsymbol{\mathcal{G}},w]} \tag{S83}$$

where the effective field-theoretic action L' is

$$\begin{aligned} L'[\mathbf{P},\boldsymbol{\mathcal{G}},w] = &\frac{1}{2}\int d\mathbf{r} \int d\mathbf{r}' \mathbf{P}(\mathbf{r}) \boldsymbol{\mathcal{T}}(\mathbf{r}-\mathbf{r}') \mathbf{P}(\mathbf{r}') + i\int d\mathbf{r}\, \mathbf{P}(\mathbf{r}) \cdot \boldsymbol{\mathcal{G}}(\mathbf{r}) - \int d\mathbf{r} \mathbf{P}(\mathbf{r}) \cdot \boldsymbol{\mathcal{E}}_0(\mathbf{r}) + i\int d\mathbf{r} w(\mathbf{r}) \\ &- \sum_S \left(\lambda'_S \int d\mathbf{r} \int d\Omega_S\, e^{i\int d\mathbf{r}'\, h_S(\mathbf{r}'-\mathbf{r}) \boldsymbol{\mu}_S \cdot \boldsymbol{\mathcal{G}}(\mathbf{r}') + i v_S w(\mathbf{r})}\right) \end{aligned} \tag{S84}$$

with $\lambda'_s = e^{\beta\mu'_s}/(4\pi\Lambda_s^3)$ being the scaled fugacity of species S.

We approximate the functional integral over $w(\mathbf{r})$ by taking the saddle-point of the action L' with respect to $w(\mathbf{r})$

$$\frac{\partial L'}{\partial w(\mathbf{r})}_{w=w^*} = 0 \tag{S85}$$

This mean-field (self-consistent field) treatment of the excluded volume effects can be justified on the basis that the fluctuation in the overall density is small in the liquid state. Noting that the saddle point value w^* is purely imaginary, we write $iw^* = \zeta^*$. Further, for a homogeneous liquid ζ^* has no position dependence. Thus, the saddle-point condition can be written as

$$\sum_S \left(\lambda'_s e^{v_S \zeta^*} v_S \int d\mathbf{r} \int d\Omega_S\, e^{i\int d\mathbf{r}'\, h_S(\mathbf{r}'-\mathbf{r}) \boldsymbol{\mu}_S \cdot \boldsymbol{\mathcal{G}}(\mathbf{r}')}\right) = 1 \tag{S86}$$

To show the connection between the grand partition function in Sec. S1 and in this section, we redefine $\mu_s = \mu'_s + v_s\zeta^*/\beta$. With the redefined chemical potential, the effective field-theoretic action L' becomes

$$\begin{aligned} L'[\mathbf{P},\boldsymbol{\mathcal{G}},w] = &\frac{1}{2}\int d\mathbf{r} \int d\mathbf{r}' \mathbf{P}(\mathbf{r}) \boldsymbol{\mathcal{T}}(\mathbf{r}-\mathbf{r}') \mathbf{P}(\mathbf{r}') + i\int d\mathbf{r}\, \mathbf{P}(\mathbf{r}) \cdot \boldsymbol{\mathcal{G}}(\mathbf{r}) - \int d\mathbf{r} \mathbf{P}(\mathbf{r}) \cdot \boldsymbol{\mathcal{E}}_0(\mathbf{r}) + i\int d\mathbf{r} w(\mathbf{r}) \\ &- \sum_S \left(\lambda_S \int d\mathbf{r} \int d\Omega_S\, e^{i\int d\mathbf{r}'\, h_S(\mathbf{r}'-\mathbf{r}) \boldsymbol{\mu}_S \cdot \boldsymbol{\mathcal{G}}(\mathbf{r}')}\right) \end{aligned} \tag{S87}$$

Comparing Eqs. (S87) with (S13), we see that $L'[\mathbf{P},\boldsymbol{\mathcal{G}},w^*] = L[\mathbf{P},\boldsymbol{\mathcal{G}}]$ results from the redefinition of the chemical potential. The grand partition function Ξ' becomes

$$\Xi' = \int \mathcal{D}\mathbf{P} \int \mathcal{D}\boldsymbol{\mathcal{G}} \int \mathcal{D}w\, e^{-L'[\mathbf{P},\boldsymbol{\mathcal{G}},w^*]} = (\text{constant}) \int \mathcal{D}\mathbf{P} \int \mathcal{D}\boldsymbol{\mathcal{G}}\, e^{-L[\mathbf{P},\boldsymbol{\mathcal{G}}]} = (\text{constant}) \times \Xi \tag{S88}$$

where the constant results from the integral $\int \mathcal{D}w$ over the functional space of $w(\mathbf{r})$. As the grand partition functions Ξ and Ξ' defer only by a constant, they are physically equivalent. Therefore, at the level of self-consistent-field treatment for the liquid density, we may absorb the effect of the incompressibility condition into the chemical potential and simply use Eq. (S7) without explicitly considering the constraint.

S3. SUPPLEMENTARY DATA

wt% of methanol	ρ (g/mL)	wt% of methanol	ρ (g/mL)	wt% of methanol	ρ (g/mL)
0	0.9982	20.0	0.9666	62.0	0.8901
0.5	0.9973	22.0	0.9636	64.0	0.8856
1.0	0.9964	24.0	0.9606	66.0	0.8810
2.0	0.9947	26.0	0.9576	68.0	0.8763
3.0	0.9930	28.0	0.9545	70.0	0.8715
4.0	0.9913	30.0	0.9514	72.0	0.8667
5.0	0.9896	32.0	0.9482	74.0	0.8618
6.0	0.9880	34.0	0.9450	76.0	0.8568
7.0	0.9864	36.0	0.9416	78.0	0.8518
8.0	0.9848	38.0	0.9382	80.0	0.8468
9.0	0.9832	40.0	0.9347	82.0	0.8416
10.0	0.9816	42.0	0.9311	84.0	0.8365
11.0	0.9801	44.0	0.9273	86.0	0.8312
12.0	0.9785	46.0	0.9235	88.0	0.8259
13.0	0.9770	48.0	0.9196	90.0	0.8204
14.0	0.9755	50.0	0.9156	92.0	0.8148
15.0	0.9740	52.0	0.9114	94.0	0.8089
16.0	0.9725	54.0	0.9072	96.0	0.8034
17.0	0.9710	56.0	0.9030	98.0	0.7976
18.0	0.9695	58.0	0.8987	100.0	0.7917
19.0	0.9680	60.0	0.8944		

TABLE S1. The density of water-methanol mixture at 20^oC used in the computation of dielectric constant of the mixture.[36]

Name	Dielectric constant[36] ε	Molar mass[44] M (g/mol)	Mass density[44] ρ (g/ml)	Solubility parameter[45] δ (MPa$^{\frac{1}{2}}$)	Molar volume (ml/mol)	Miscibility with water[37]	Miscibility with methanol[37]	Miscibility with cyclohexane[37]
Acetic Acid	6.2	60.052	1.049	20.7	57.247	Y	Y	Y
Acetone	21.01	58.079	0.791	20.2	73.425	Y	Y	Y
Acetonitrile	36.64	41.052	0.786	24.3	52.229	Y	Y	N
Benzene	2.2825	78.112	0.874	18.8	89.373	N	Y	Y
1-Butanol	17.84	74.122	0.810	23.3	91.509	N	Y	Y
Butyl Acetate	5.07	116.158	0.880	17.4	131.998	N	Y	Y
Tetrachloromethane	2.2379	153.823	1.594	17.6	96.501	N	Y	Y
Trichloromethane	4.8069	119.378	1.492	19.0	80.012	N	Y	Y
Cyclohexane	2.0243	84.160	0.779	16.8	108.036	N	N	N.A.
1,1-Dichloroethane	10.36	98.959	1.180	20	83.864	N	Y	Y
Dichloromethane	8.997	84.933	1.325	19.8	64.100	N	Y	Y
Diethyl Ether	4.2666	74.122	0.713	15.1	103.958	N	Y	Y
Diisopropyl Ether	3.805	102.175	0.725	14.1	140.931	N	Y	Y
Dimethylformamide	38.25	73.094	0.944	24.8	77.430	Y	Y	N
Dimethyl Sulfoxide	47.24	78.133	1.100	24.5	71.030	Y	Y	N
1,4-Dioxane	2.2189	88.105	1.034	20.5	85.208	Y	Y	Y
Ethanol	25.3	46.068	0.789	26.0	58.388	Y	Y	Y
Ethyl Acetate	6.0814	88.105	0.902	18.6	97.677	N	Y	Y
Heptane	1.9209	100.202	0.684	15.1	146.494	N	N	Y
Hexane	1.8865	86.175	0.659	14.9	130.766	N	N	Y
Isooctane	1.869	114.229	0.692	14.1	165.071	N	N	Y
Methanol	33	32.042	0.791	29.6	40.508	Y	N.A.	N
2-Butanone	18.56	72.106	0.805	19.0	89.573	N	Y	Y
Pentane	1.8371	72.149	0.626	14.3	115.254	N	N	Y
1-Propanol	20.8	60.095	0.804	24.3	74.745	Y	Y	Y
Tetrahydrofuran	7.563	72.106	0.889	18.6	81.109	Y	Y	Y
Toluene	2.387	92.138	0.867	18.2	106.272	N	Y	Y
Trichloroethylene	3.462	131.388	1.460	18.8	89.992	N	Y	Y
Water	80.1	18.015	1.000	47.9	18.015	N.A.	Y	N
o-Xylene	2.562	106.165	0.879	18.0	120.779	N	Y	Y

TABLE S2. Parameters for the solvents for the computation of Figure 3 in the main manuscript. "Y"/"N" indicates a liquid that is miscible/immiscible with the liquid mentioned in the header.

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